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2004 J. Phys.: Condens. Matter 16 1245

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INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **16** (2004) 1245–1267 PII: S0953-8984(04)64860-0

Spatial dispersion in incommensurately modulated insulators

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Received 16 June 2003
Published 13 February 2004
Online at stacks.iop.org/JPhysCM/16/1245 (DOI: 10.1088/0953-8984/16/8/009)

Abstract

Peculiarities of the non-local responses of spatially inhomogeneous insulating media to electromagnetic waves are considered within a phenomenological approach. In the case of a weak spatial dispersion (SD), a 'quasi-local' constitutive equation is derived, which allows one to isolate the effects of SD in relation to non-uniformities of the wavefield and the material properties of the medium. The symmetry of the material tensors is clarified. The material contribution to the SD suggests a novel mechanism for optical activity. The SD in crystalline media is described in terms of dielectric permittivity dependent upon a single argument which is a specific combination of the light wavevector and the reciprocal lattice vectors. The conditions and materials for which the relevant effects can be notable are pointed out. The character of the normal light wave polarization in dielectric crystals of the A_2BX_4 group with incommensurately modulated superstructure is analysed and the effective crystal optical parameters are derived. It is shown that the first-order material SD effects associated with the bulk macroscopic properties of these crystals could be of practical importance.

1. Introduction

Spatial dispersion (SD) in the propagation of electromagnetic waves in solids, liquids, plasmas etc is an analogue of ordinary (time or frequency) dispersion [1] on the spatial scale. The insulating medium is said to be spatially dispersive if its optical response is non-local, i.e. the electric displacement $\mathbf{D}(\mathbf{r})$ of an electromagnetic wave at a given point $\mathbf{r}$ depends upon the electric field $\mathbf{E}(\mathbf{r})$ at this point and the field values $\mathbf{E}(\mathbf{r}')$ in some vicinity of the point [2]. Then the non-local functional relation between $\mathbf{D}$ and $\mathbf{E}$ (or the so-called constitutive relation, abbreviated hereafter as CR) written in the spatial domain for a linear anisotropic continuous medium in the frequency region far from resonances takes the form [2]

$$D_i(\mathbf{r}) = \int \epsilon_{ij}(\mathbf{r}, \mathbf{r}') E_j(\mathbf{r}')\, \mathrm{d}\mathbf{r}' \tag{1}$$

where the integration is in fact performed over the volume of the medium and the summation convention over repeated Cartesian indices is used. The kernel $\epsilon_{ij}(\mathbf{r}, \mathbf{r}')$ of the integral operator

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in the rhs of equation (1) represents the response function of the medium and the quantities **E** and **D** entering there are not necessarily macroscopically averaged. Fourier transforming (1) for spatially homogeneous media yields an optical dielectric tensor ε_{ij} dependent on the light wavevector $\mathbf{k}$ ($D_i(\mathbf{k}) = \varepsilon_{ij}(\mathbf{k})E_j(\mathbf{k})$—see, e.g., [2, 3]), which is usually believed to represent the most general (and quite equivalent to equation (1)) formulation of SD for any media.

It is well known that the SD modifies notably optical properties of the medium, producing, in particular, an optical activity (or gyration) effect. According to traditionally accepted formulations [4, 5], the optical activity originates from the first-order SD corrections to a common 'local' optics and is understood as a result of finiteness of the electromagnetic wavelength λ with respect to the molecule dimensions or the crystal lattice parameters a. The effect has therefore a typical order of magnitude a/λ and arises under circumstances when the electric field non-uniformity on the a scale cannot be neglected.

Since the inhomogeneity of material media on the microscopic (atomic) scales does not in fact contribute to the optical response (see the analysis by Agranovich and Ginzburg [2]), while the scales of spatial variations of the macroscopic properties of the media are typically much larger than the light wavelength, the consideration of SD has been so far restricted mainly to homogeneous media. Unfortunately, this approximate approach leaves out of consideration the optical phenomena that can, in principle, occur in the media which seem to be certainly inhomogeneous (solid state systems possessing superstructures of different physical nature, non-uniform waveguides and plasmas, liquid crystals etc). Therefore, serious questions remain in the theory of SD in matters related to its manifestations in spatially inhomogeneous materials. In our view, the expected weakness of the above mentioned phenomena cannot be regarded as absolutely definite.

In this respect, the analysis of light propagation in bounded media, the limiting case of inhomogeneous ones (see the results [6–8] and, especially, [9–11]), is very interesting. It requires taking into account spatial variations of the dielectric parameters in the surface layer, in order to obtain correct energetic relationships within the phenomenological electrodynamics of gyrotropic media. In our opinion, however, the approach used in [9–11] is not sufficiently general, while the corresponding results require a proper physical interpretation. Additional interest in the SD in inhomogeneous media was stimulated by the works [12–15], in which the formulation of the crystal optics of the insulating materials possessing IC superstructure is fundamentally based on the assumption of their non-uniformity on semi-macroscopic scales.

The aim of our study is to analyse the SD in inhomogeneous media and its consequences for the observable, macroscopic crystal optical properties. We present simple physical considerations concerning the peculiarities of SD in macroscopically inhomogeneous media, particularly crystalline ones. We wish to show that a new kind of SD phenomenon, described by the same equation (1), can be distinguished for these media, which produces optical activity with a different origin. The outline of the present paper is as follows. In section 2 we begin with some important results of the phenomenological theory of SD in homogeneous continuous insulating media. The consideration in section 3 is generalized towards inhomogeneous media and the mechanisms for the optical gyration in those media are clarified. Section 4 is devoted to the description of the SD in crystalline media, in particular crystals possessing the incommensurate (IC) superstructure. Section 5 deals with the quantitative analysis of the influence of non-locality of the optical response and the mesoscopic structural inhomogeneity of the IC crystals on the character of the electromagnetic wave polarization in those crystals. The crystal optical characteristics of centrosymmetric IC phases are obtained there and the possibility for experimental observation of first-order SD effects is discussed. Finally, the conclusions are drawn in section 6.

2. Spatial dispersion in homogeneous continuous media

We start with discussing the most important points related to SD in homogeneous media, which will be later used in the description of more complicated cases. This brief discussion is also justified by the inconsistencies that occur sometimes in the presentation of the subject. First of all, the medium has to be regarded as infinite; for any boundary, at which material parameters change abruptly, represents a partial case of inhomogeneity. The properties of a homogeneous, infinitely extended continuous medium should be invariant with respect to translations, including infinitesimal ones, and so we have in (1)

$$D_i(\mathbf{r}) = \int \epsilon_{ij}(\mathbf{r}-\mathbf{r}')E_j(\mathbf{r}')\,\mathrm{d}\mathbf{r}'. \tag{2}$$

Note that a further assumption of a dependence of the kernel ϵ_{ij} on the modulus $|\mathbf{r}-\mathbf{r}'|$ would have meant the presence of an inversion centre in the medium (equivalence of all directions to the opposite ones) and the absence of gyrotropy[1]. On the other hand, this is also evident because a response function even in $\mathbf{r}-\mathbf{r}'$ would forbid the property (see below).

Despite the formally infinite integration limits in (2), it is understood from the simplest physical reasoning that the optical processes at notably distant points cannot be concerned with each other. Therefore, the kernel $\epsilon_{ij}(\mathbf{r}-\mathbf{r}')$ remains non-zero only for argument values not too much larger than a characteristic 'non-locality radius' a_s, which determines the importance of the non-locality phenomena. In practice, a_s is equated to a free path length of particles or the Debye radius in plasmas [3] and to molecule or ionic group dimensions, or the interatomic distances (the lattice parameters) a in a condensed matter [2]. If non-localized collective excitations are present in crystals, a_s can sometimes exceed typical dimensions of the unit cell [16].

When we are allowed, under some circumstances, to consider a_s as being infinitely small, we can put $\epsilon_{ij}(\mathbf{r}-\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}')\varepsilon_{ij}^{\mathrm{NSD}}$ in (2), where $\delta(\mathbf{r})$ is the Dirac delta function, and the local optics limit is recovered:

$$D_i(\mathbf{r}) = \varepsilon_{ij}^{\mathrm{NSD}}E_j(\mathbf{r}) \tag{3}$$

which describes spatially non-dispersive medium. The situation could be compared with the case of a spatially dispersive medium responding to a uniform field ($\mathbf{E}(\mathbf{r}') = \mathbf{E}(\mathbf{r}) = \mathbf{E}$). Then on the basis of formula (2) we obtain

$$D_i = \varepsilon_{ij}^{\mathrm{SD}}E_j \tag{4}$$

where $\varepsilon_{ij}^{\mathrm{SD}} = \int \epsilon_{ij}(\mathbf{r}-\mathbf{r}')\,\mathrm{d}\mathbf{r}'$. The cases of equation (3) for a constant field and (4) differ only in the definitions of the material parameters. Thus, both non-locality of the response ($a_\mathrm{s} \neq 0$) and non-uniformity of the external field are necessary for the SD to manifest itself in the homogeneous medium. The above mentioned non-uniformity is described by characteristic scales of essential changes in the field, in particular by the wavelength λ for electromagnetic waves. Thus the importance of SD is determined by the ratio a_s/λ.

If $a_\mathrm{s}/\lambda \ll 1$, the field $\mathbf{E}(\mathbf{r}')$ may be expanded in a series in the vicinity of the point $\mathbf{r}$. Confining ourselves to several lowest-order expansion terms, we have a 'quasi-local' CR, instead of (2):

$$D_i(\mathbf{r}) = \varepsilon_{ij}E_j(\mathbf{r}) + \gamma_{ijl}^{(1)}\nabla_l E_j(\mathbf{r}) + \gamma_{ijlm}^{(2)}\nabla_l\nabla_m E_j(\mathbf{r}) + \cdots \tag{5}$$

[1] The response function of an isotropic, non-gyrotropic medium obtained within a specific model in section 10 in [2] depends only on $|\mathbf{r}-\mathbf{r}'|$.

where

$$\begin{aligned}\varepsilon_{ij} &= \int \epsilon_{ij}(\mathbf{r}-\mathbf{r}')\,\mathrm{d}\mathbf{r}',\\ \gamma^{(1)}_{ijl} &= \int \epsilon_{ij}(\mathbf{r}-\mathbf{r}')(\mathbf{r}'-\mathbf{r})_l\,\mathrm{d}\mathbf{r}',\\ \gamma^{(2)}_{ijlm} &= \int \epsilon_{ij}(\mathbf{r}-\mathbf{r}')(\mathbf{r}'-\mathbf{r})_l(\mathbf{r}'-\mathbf{r})_m\,\mathrm{d}\mathbf{r}';\end{aligned} \tag{6}$$

ε_{ij} is the dielectric permittivity tensor which does not account for the SD, while $\gamma^{(1)}_{ijl}$ and $\gamma^{(2)}_{ijlm}$ describe the SD effects of the first (gyrotropy) and the second orders, respectively.

Let us analyse some symmetry properties of the response functions $\epsilon_{ij}(\mathbf{r},\mathbf{r}')$ and $\epsilon_{ij}(\mathbf{r}-\mathbf{r}')$ and the material tensors in formula (5). The fundamental Onsager symmetry principle for the kinetic coefficients [5] applied to the dielectric permittivity of spatially dispersive media requires that the condition

$$\int \mathbf{E}^{(1)}(\mathbf{r})\mathbf{D}^{(2)}(\mathbf{r})\,\mathrm{d}\mathbf{r} = \int \mathbf{E}^{(2)}(\mathbf{r})\mathbf{D}^{(1)}(\mathbf{r})\,\mathrm{d}\mathbf{r} \tag{7}$$

be satisfied for arbitrary electric fields $\mathbf{E}^{(1)}$ and $\mathbf{E}^{(2)}$ and the corresponding displacements $\mathbf{D}^{(1)}$, $\mathbf{D}^{(2)}$. With (1) and (2), this leads to

$$\epsilon_{ij}(\mathbf{r},\mathbf{r}') = \epsilon_{ji}(\mathbf{r}',\mathbf{r}), \qquad \epsilon_{ij}(\mathbf{r}-\mathbf{r}') = \epsilon_{ji}(\mathbf{r}'-\mathbf{r}). \tag{8}$$

One can note that formula (7) reduces to the condition of self-conjugate (Hermitian) character of the 'overall' dielectric permittivity $\hat{\varepsilon}$, which gives the electric displacement according to $\mathbf{D} = \hat{\varepsilon}\mathbf{E}$, if the scalar product of vectors is only defined through the 'natural' relation $(\mathbf{E}^{(1)},\mathbf{E}^{(2)}) \equiv \int \mathbf{E}^{(1)}(\mathbf{r})\mathbf{E}^{(2)}(\mathbf{r})\,\mathrm{d}\mathbf{r}$ in a real linear vector space. In order for the integral operator $\hat{\varepsilon}$, which acts according to (1) or (2), not to take the vectors out of the linear space, its kernel $\epsilon_{ij}(\mathbf{r},\mathbf{r}')$ (or $\epsilon_{ij}(\mathbf{r}-\mathbf{r}')$) should be real:

$$\epsilon_{ij}(\mathbf{r},\mathbf{r}') = [\epsilon_{ij}(\mathbf{r},\mathbf{r}')]^*, \qquad \epsilon_{ij}(\mathbf{r}-\mathbf{r}') = [\epsilon_{ij}(\mathbf{r}-\mathbf{r}')]^*. \tag{9}$$

In other words, the Onsager principle would result in Hermitian dielectric permittivity, provided that the condition (9) is fulfilled. In particular, the symmetry of the kernel of the 'scalar' Fredholm operator $\hat{\varepsilon}\mathbf{E}(\mathbf{r}) = \int \epsilon(\mathbf{r},\mathbf{r}')\mathbf{E}(\mathbf{r}')\,\mathrm{d}\mathbf{r}'$ with respect to interchanging coordinates ($\epsilon(\mathbf{r},\mathbf{r}') = \epsilon(\mathbf{r}',\mathbf{r})$) makes the operator Hermitian whenever the kernel is real.

On the other hand, the condition of absence of energy losses $\int \mathbf{E}(\mathbf{r})[\mathbf{D}(\mathbf{r})]^*\,\mathrm{d}\mathbf{r} = \int [\mathbf{E}(\mathbf{r})]^*\mathbf{D}(\mathbf{r})\,\mathrm{d}\mathbf{r}$ [5] for the electric component of a harmonic wave $\sim \exp \mathrm{i}\omega t$, which is fulfilled for transparent media, just reduces to (9). This ensures that the dielectric permittivity of the lossless medium is Hermitian.

On the basis of the Onsager symmetry for the real kernel $\epsilon_{ij}(\mathbf{r}-\mathbf{r}')$ and the definitions (6), the known properties of the material tensors in (5) hold true for the homogeneous media:

$$\varepsilon_{ij} = \varepsilon_{ji} \qquad \gamma^{(1)}_{ijl} = -\gamma^{(1)}_{jil} \qquad \gamma^{(2)}_{ijlm} = \gamma^{(2)}_{jilm}, \tag{10a}$$

$$\varepsilon_{ij} = \varepsilon^*_{ij} \qquad \gamma^{(1)}_{ijl} = [\gamma^{(1)}_{ijl}]^* \qquad \gamma^{(2)}_{ijlm} = [\gamma^{(2)}_{ijlm}]^*. \tag{10b}$$

In deriving formulae (10), we have taken into account that $\epsilon_{ij}(\mathbf{r}-\mathbf{r}')$ contains components both even and odd in $\mathbf{r}-\mathbf{r}'$. They determine the tensors ε_{ij}, $\gamma^{(2)}_{ijlm}$ and the gyration tensor $\gamma^{(1)}_{ijl}$, respectively. As a result, the absence of the odd component in $\epsilon_{ij}(\mathbf{r}-\mathbf{r}')$ and the absence of the gyration effect are in fact equivalent. Besides this, the trivial property $\gamma^{(2)}_{ijlm} = \gamma^{(2)}_{ijml}$ holds for the $\gamma^{(2)}_{ijlm}$ tensor.

3. Spatial dispersion in inhomogeneous media

3.1. 'Quasi-local' constitutive relation

In the case of inhomogeneity, including that inevitably present in every material medium on the microscopic level, the dielectric response should additionally depend on the coordinates $\mathbf{r}$ and $\mathbf{r}'$ separately, besides the dependence on the $\mathbf{r}-\mathbf{r}'$ argument mentioned above [2, 3, 6]. It has usually been supposed that the response function takes the form $\epsilon_{ij}(\mathbf{r}-\mathbf{r}', \mathbf{r})$ [17] or $\epsilon_{ij}(\mathbf{r}-\mathbf{r}', \mathbf{r}')$ (see the consideration of SD in crystals [2], chapter 2, section 13). In this work we assume the additional dependence to be 'symmetric' in $\mathbf{r}$ and $\mathbf{r}'$. In other words, the kernel ϵ_{ij} should depend on $\mathbf{r}'$ (at a fixed $\mathbf{r}$) in the same way as on $\mathbf{r}$ (at a fixed $\mathbf{r}'$), with the corresponding interchange of the integration variable in (1). Further consideration shows that this assumption has a close relation to the Onsager symmetry (formulae (7) and (8)). Note also that Johnson and Rimbey [8] worked in terms of the additional argument $z+z'$ in the case of a semi-infinite medium with a plane boundary perpendicular to the z axis, in agreement with our assumption. Of course, the dependence of the kernel on $\mathbf{r}-\mathbf{r}'$ has all the features discussed above.

Hence, we will represent the integral kernel of CR (1) for inhomogeneous media in the form

$$\epsilon_{ij}(\mathbf{r}, \mathbf{r}') = \epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2) \tag{11}$$

where for the sake of concreteness the arguments $\mathbf{r}_1$ and $\mathbf{r}_2$ are defined as

$$\mathbf{r}_1 = \mathbf{r}-\mathbf{r}', \qquad \mathbf{r}_2 = \tfrac{1}{2}(\mathbf{r}+\mathbf{r}'). \tag{12}$$

Here the variables $\mathbf{r}_1$ and $\mathbf{r}_2$ can be regarded as independent, inasmuch as the functional dependences of the kernel on $\mathbf{r}_1$ and $\mathbf{r}_2$ represent different physical effects. That is, the former reflects non-locality of the response, while the latter describes the inhomogeneity of the optical properties, e.g., a spatial variation of the non-locality radius. One can further prove that other choices of constants α and β in the more general definitions $\mathbf{r}_1 = \alpha(\mathbf{r}-\mathbf{r}')$ and $\mathbf{r}_2 = \beta(\mathbf{r}+\mathbf{r}')$ (with finite non-zero constants α and β) would not cause violation of the main conclusions of this work (see appendix A).

Let us introduce a characteristic dimension λ_m where the properties of the medium change appreciably (say, a spatial period in the case of periodic media). If the λ_m parameter is not infinitely large as compared to the λ value, the SD can no longer be attributed to non-uniformity of the electromagnetic wave alone. Consistent consideration requires taking into account spatial variations of the material parameters of the medium itself. In other words, whenever the non-locality scales a_s, on which the response to the wave is formed, are not negligibly small with respect to λ and λ_m, then both the wavefield and the properties of the medium are to be considered as variable on those scales. It is seen from (1), (11) and (12) that the displacement $\mathbf{D}(\mathbf{r})$ is determined not only by the field $\mathbf{E}(\mathbf{r})$ and the local material parameters $\epsilon_{ij}(\mathbf{r}_1, \mathbf{r})$ at the point $\mathbf{r}$ (i.e., the values $\epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2)$ taken at $\mathbf{r}' = \mathbf{r}$ in the argument $\mathbf{r}_2$), but also the fields $\mathbf{E}(\mathbf{r}')$ and the parameters $\epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2)$ in some vicinity of that point.

Let us now proceed to a quantitative description of these phenomena. When the ratios a_s/λ and $a_\mathrm{s}/\lambda_\mathrm{m}$ are negligible, we have $\epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2) = \delta(\mathbf{r}_1)\varepsilon_{ij}^{\mathrm{NSD}}(\mathbf{r}_2)$ from (11) and (12) and so pass to the local CR (cf equation (3)),

$$D_i(\mathbf{r}) = \varepsilon_{ij}^{\mathrm{NSD}}(\mathbf{r})E_j(\mathbf{r}). \tag{13}$$

When the SD is weak (a_s/λ, $a_\mathrm{s}/\lambda_\mathrm{m} \ll 1$), the response function (11) and the field in (1) may to a sufficient degree of accuracy be represented by several lowest-order terms of their Taylor series. After transformations (see appendix A), the corresponding expansion is as follows:

$$\begin{aligned} D_i(\mathbf{r}) = {} & \varepsilon_{ij}(\mathbf{r})E_j(\mathbf{r}) + \gamma_{ijl}^{(1)}(\mathbf{r})\nabla_l E_j(\mathbf{r}) + \tfrac{1}{2}[\nabla_l \gamma_{ijl}^{(1)}(\mathbf{r})]E_j(\mathbf{r}) + \gamma_{ijlm}^{(2)}(\mathbf{r})\nabla_l\nabla_m E_j(\mathbf{r}) \\ & + [\nabla_l \gamma_{ijlm}^{(2)}(\mathbf{r})]\nabla_m E_j(\mathbf{r}) + \tfrac{1}{4}[\nabla_l\nabla_m \gamma_{ijlm}^{(2)}(\mathbf{r})]E_j(\mathbf{r}) + \cdots \end{aligned} \tag{14}$$

where the ∇s affect the nearest square brackets only and the material tensors of the inhomogeneous medium are defined according to[2]

$$\begin{aligned}
\varepsilon_{ij}(\mathbf{r}) &= 2\int \epsilon_{ij}(\mathbf{r}-\mathbf{r}',\mathbf{r})\,\mathrm{d}\mathbf{r}',\\
\gamma^{(1)}_{ijl}(\mathbf{r}) &= 4\int \epsilon_{ij}(\mathbf{r}-\mathbf{r}',\mathbf{r})(\mathbf{r}'-\mathbf{r})_l\,\mathrm{d}\mathbf{r}',\\
\gamma^{(2)}_{ijlm}(\mathbf{r}) &= 4\int \epsilon_{ij}(\mathbf{r}-\mathbf{r}',\mathbf{r})(\mathbf{r}'-\mathbf{r})_l(\mathbf{r}'-\mathbf{r})_m\,\mathrm{d}\mathbf{r}'.
\end{aligned} \tag{15}$$

It is seen from (14) that the SD phenomena in a dielectric medium characterized by a non-local response are related to inhomogeneities of both the electric field of the electromagnetic wave and the optical parameters of the medium. In the following, we shall conventionally refer to these phenomena as the SD attributed to the wavefield (or simply *field* SD) and the SD attributed to the material (or *material* SD), respectively. The second-order and higher-order SD effects contain additionally terms of a 'mixed' type ($[\nabla_l\gamma^{(2)}_{ijlm}]\nabla_m E_j$ etc). The field and the material SDs are hardly separable in general (see equation (1)), though the only exception concerns a practical case of a weak first-order SD. It is worth noticing that the material SD effect should manifest itself in inhomogeneous media even if the latter are responding to a spatially uniform external field. This would essentially modify the electric displacement when compared to the case of a non-dispersive medium (cf the situation described by equation (4)).

3.2. Symmetry of the material tensors

In order to analyse the symmetry of spatially dependent tensors in (14), it would be more convenient to employ the initial formulation of the Onsager principle in the form of a 'cross-energy' relationship (7). Using (7) and (14), we obtain

$$\begin{aligned}
E^{(1)}_i E^{(2)}_j[(\varepsilon_{ij}-\varepsilon_{ji}) &+ \tfrac{1}{2}(\gamma^{(1)}_{ijl}+\gamma^{(1)}_{jil}) + \tfrac{1}{4}(\gamma^{(2)}_{ijlm}-\gamma^{(2)}_{jilm})]\\
&+ E^{(1)}_i[(\gamma^{(1)}_{ijl}+\gamma^{(1)}_{jil}) + \nabla_m(\gamma^{(2)}_{ijlm}-\gamma^{(2)}_{jilm})]\nabla_l E^{(2)}_j\\
&+ E^{(1)}_i[\gamma^{(2)}_{ijlm}-\gamma^{(2)}_{jilm}]\nabla_l\nabla_m E^{(2)}_j = 0,
\end{aligned} \tag{16}$$

where the trivial property $\gamma^{(2)}_{ijlm} = \gamma^{(2)}_{ijml}$ (see formula (15)) is taken into account. Since the functions $\mathbf{E}^{(1)}$ and $\mathbf{E}^{(2)}$ entering (16) are arbitrary, formulae (10*a*) are also valid for the inhomogeneous medium; i.e., the tensors ε_{ij} and $\gamma^{(2)}_{ijlm}$ are symmetric and $\gamma^{(1)}_{ijl}$ antisymmetric under interchange of the indices i and j. If the terms related to the material SD were absent in the 'quasi-local' CR, it would not agree with the Onsager principle. Thus we must have arrived at a requirement of the type of (16), from which there follows a need for either putting $\gamma^{(1)}_{ijl} = \gamma^{(2)}_{ijlm} = \cdots =$ constant or redefining the material tensors in a complicated way, which might be implicitly reduced to formula (14).

A similar situation would arise if the numerical coefficients of the material SD contributions in (14) were altered. According to the general regularity that follows from formula (A.7) of appendix A, the coefficients of 'purely material' contributions to the pth-order SD are 2^p times less than those of the 'purely field' contributions. This is a direct consequence of our assumption concerning the $\mathbf{r}-\mathbf{r}'$ symmetry in the additional coordinate dependence of

[2] The limiting transition to the case of a homogeneous medium with the tensor definitions (formulae (6)) needs some caution. So, $\varepsilon_{ij}(\mathbf{r})$ is determined not only by the first term in the expansion of the kernel $\epsilon_{ij}(\mathbf{r}_1,\mathbf{r}_2)$ (i.e., $\epsilon_{ij}(\mathbf{r}_1,\mathbf{r})$) but also all the following terms (see formula (A.1)). For instance, the second term $(1/2)\nabla_l[\epsilon_{ij}(\mathbf{r}_1,\mathbf{r})](\mathbf{r}'-\mathbf{r})_l = (1/2)\nabla_l[\epsilon_{ij}(\mathbf{r}_1,\mathbf{r})(\mathbf{r}'-\mathbf{r})_l] + (1/2)\epsilon_{ij}(\mathbf{r}_1,\mathbf{r})$ gives the additional contribution $(1/2)\epsilon_{ij}(\mathbf{r}_1,\mathbf{r})$. Since all the spatial derivatives of $\epsilon_{ij}(\mathbf{r}_1,\mathbf{r})$ are zero for the homogeneous medium, the above mentioned contributions are absent and the coefficient '2' in the definition (15) of $\varepsilon_{ij}(\mathbf{r})$ transforms to unity in the definition (6) of ε_{ij}.

the kernel of the integral CR for inhomogeneous media. Thus, the latter assumption may be regarded as equivalent to the requirement of Onsager symmetry of the optical response. The partial case of the above mentioned regularity (the presence of the coefficient $1/2$ for the term $(\nabla_l \gamma^{(1)}_{ijl}) E_j$ entering the CR for the bounded gyrotropic media) has long been disputed in the literature and was proved by Agranovich and Yudson [9] (see also [2, 10, 11]). Strictly speaking, the motivation of the authors of [9, 10] for introducing the first-order spatial derivatives of the gyration tensor into the quasi-local CR just consisted in the desire to formally adjust the CR to the requirements of the Onsager principle and the balance conditions for the electromagnetic energy flow through the boundary of the medium. Unfortunately, this was accompanied with neither discussion of the phenomena associated with the material SD nor a relevant analysis of its physical consequences.

Using the condition for transparency of the medium (see section 2), together with (10*a*), leads again to formulae (10*b*). Thus, all the tensors in (14) should be real and, with formula (15), the same applies to the $\epsilon_{ij}(\mathbf{r} - \mathbf{r}', \mathbf{r})$ function. In other words, the symmetry of the material tensors describing the optical properties of inhomogeneous media remains unchanged as compared with the case of homogeneous media. In particular, when the absorption is absent, the 'overall' dielectric permittivity tensor $\hat{\varepsilon}$ defined formally via the relation $\mathbf{D} = \hat{\varepsilon}\mathbf{E}$ is Hermitian, thus objecting to a groundless statement in the recent work [15]. It is worthwhile noting that, under the conditions of spatial dependence of the material parameters, the inversion symmetry of the medium does not necessarily lead to disappearance of the odd-rank tensors in (14). So, we have $\gamma^{(1)}_{ijl}(\mathbf{r}) = -\gamma^{(1)}_{ijl}(-\mathbf{r})$ for the gyration tensor under the action of the inversion centre, in clear contrast to the condition $\gamma^{(1)}_{ijl} = -\gamma^{(1)}_{ijl}$ that forbids gyrotropy for homogeneous medium. This general conclusion agrees well with the result of Meekes and Janner [12] for the IC crystals obtained more rigorously within the superspace symmetry approach.

Finally, we note that an attempt to relate the non-local response function and the tensors $\varepsilon_{ij}(\mathbf{r})$ and $\gamma^{(1)}_{ijl}(\mathbf{r})$ in the CR [9] for the transition layer at the gyrotropic medium surface has been made in [11]. Using the notation adopted in this work, the author of [11] has defined the material tensors as $\varepsilon_{ij}(\mathbf{r}) = \int \epsilon_{ij}(\mathbf{r}, \mathbf{r}')\,\mathrm{d}\mathbf{r}'$ and $\gamma^{(1)}_{ijl}(\mathbf{r}) = \int \epsilon_{ij}(\mathbf{r}, \mathbf{r}')(\mathbf{r}' - \mathbf{r})_l\,\mathrm{d}\mathbf{r}'$. On the basis of (11), (A.1) and the results of appendix A, one can conclude that $\varepsilon_{ij}(\mathbf{r})$ and $\gamma^{(1)}_{ijl}(\mathbf{r})$ include contributions from the higher-order SD and so are asymmetric in the indices i and j. Therefore, the definitions turn out to be both inconvenient and inconsistent, and the same applies to the final CR for the gyrotropic medium (see formula (12) in [11]).

3.3. The effects related to the material spatial dispersion

Let us discuss the possible physical consequences of the material SD. First of all, we recall that the common understanding of the SD in non-magnetic media as phenomena associated with a dependence of the electric polarization (or displacement) at a given point on the electric field values in the vicinity of that point (see, e.g., [2, 3, 5]) should be admitted to be a restricted one. It is interesting that the results [9–11, 18, 19] (see also [2], section 3) have still not hampered such reductive interpretations, since they have only been linked with the transition layer at the surface of a gyrotropic medium. According to our results, the notion of SD should be generalized to one embracing the optical phenomena which appear due to non-locality of the response and manifest themselves under conditions of sufficiently inhomogeneous wavefield and/or material properties of the medium.

As mentioned in section 1, SD could be compared with the ordinary (time or frequency) dispersion, which has a decisive effect in optics and is caused by inertiality of the electric polarization of the medium (see [1]). It is well known that the displacement at a given time t is

determined in a non-local fashion by both the field at that particular instant of time and the fields at the preceding times $t' < t$, provided that the relaxation time τ_s for the medium polarization process is not negligibly small when compared with the field period T. More strictly, we have $D_i(t) = \int_{-\infty}^{t} \epsilon_{ij}(t, t')E_j(t')\,dt'$. In the case of a stationary medium (a constant τ_s value, $\epsilon_{ij}(t, t') = \epsilon_{ij}(t - t')$), this leads to the material relation $D_i(\omega) = \varepsilon_{ij}(\omega)E_j(\omega)$ in terms of the Fourier components, and a dependence of the material parameters on the frequency ω of the electromagnetic wave. In this context, the material SD has an evident analogue in the time dispersion phenomena occurring in weakly non-stationary media, under the condition of finiteness of characteristic times T_m required for notable variations of the properties of the medium ($\tau_s/T_m \neq 0$), e.g., due to non-linear optical effects appearing under the action of the other intense electromagnetic wave. However, further elaboration of the analogy could hardly be efficient, for the non-stationary media themselves have so far been given little consideration (see [5], section 61).

One can grasp some consequences of the material SD even without a detailed quantitative analysis. First, we need to come to terms with the definitions. A comparison with the simplest case of macroscopically homogeneous, transparent crystals manifesting first-order SD would be useful in this regard. In fact, two different definitions of optical gyration may be utilized for the latter objects (see [2, 4, 5, 20, 21]). One can reasonably postulate that the crystal is optically active if the corresponding CR includes a non-zero tensor $\gamma^{(1)}_{ijl}$. Instead of this definition, dealing with the material parameters of the crystal, one can, quite equivalently, define the gyration 'physically', making use of the fact that the waves propagating in that crystal are elliptically (in particular cases, circularly) polarized [20].

Let us assume that the plane electromagnetic wave with the wavevector $\mathbf{k} = \mathbf{e}_z k$ ($e_i = 1$, $i = x, y, z$ being the principal axes of the optical indicatrix ellipsoid) and the electric field $\mathbf{E} = \mathbf{e}_x E_x \exp \mathrm{i}(\omega t + kz)$ in the complex representation is excited in such a crystal, which possesses, e.g., an orthorhombic symmetry. Since the overall dielectric permittivity is $\hat{\varepsilon}_{ij} = \varepsilon_{ij} + \mathrm{i}\gamma^{(1)}_{ijl}k_l$ (with $\varepsilon_{ij} = 0$ at $i \neq j$—see formula (5)), the displacement field would be polarized elliptically ($\mathbf{D} = [\mathbf{e}_x\varepsilon_{11} - \mathrm{i}\mathbf{e}_y\gamma^{(1)}_{123}k]E_x \exp \mathrm{i}(\omega t + kz)$). Here the phase retardation $\pi/2$ between the components D_x and D_y and so also the elliptical character of the polarization are due to antisymmetry of the $\gamma^{(1)}_{ijl}$ tensor and imaginary second term ($\gamma^{(1)}_{ij3}\nabla_z = \mathrm{i}\gamma^{(1)}_{ij3}k$) in the $\hat{\varepsilon}_{ij}$ tensor. The crystal is, of course, optically active.

Now let the said wave be excited in the inhomogeneous (for the sake of definiteness, periodic along the z axis) medium, characterized by the period $\lambda_m = 2\pi/q$ and the tensor $\hat{\varepsilon}_{ij}(\mathbf{r}) = \varepsilon_{ij} + \gamma^{(1)}_{ijl}(\mathbf{r})\nabla_l + (1/2)[\nabla_l\gamma^{(1)}_{ijl}(\mathbf{r})]$. It is well known [21] that the typical solutions for the electromagnetic waves may be represented as non-uniform Bloch-type waves $\mathbf{D} = [\mathbf{e}_x\varepsilon_{11}E_x + \mathbf{e}_y(A^+_y \exp \mathrm{i}qz + A^-_y \exp(-\mathrm{i}qz))] \exp \mathrm{i}(\omega t + kz)$, where $A^{\pm}_y$ are the real amplitude coefficients determined by E_x and the off-diagonal components of $\hat{\varepsilon}_{ij}(\mathbf{r})$. The waves turn out to be elliptically polarized in general, the maximum ellipticity being achieved at the points where $qz = \pm\pi/2$. Starting from the above, it is expedient to qualify the medium as optically active, although it could not be completely fitted into the common definitions of gyration for homogeneous media from the viewpoints of the 'quasi-local' CR and the normal wave polarization (see also the discussion in [22]).

Hence, the first-order material SD leads to an apparent gyration, which can be, in principle, detected experimentally (cf the long-accepted formulations of gyration given by workers in the field [2, 4, 5, 20, 21]). The effect should be evaluated via the ratio a_s/λ_m, not a_s/λ. As far as we know, the relevant physical mechanism for the gyration associated with the finiteness of the characteristic dimensions of inhomogeneity of the material—in particular, the superstructure periods or any other periods imposed by non-uniform external influences—has never been mentioned in the literature.

As far as the microscopic wavefields and the material properties are concerned, formula (14) is the only relation consistent with the general symmetry requirements. On passing to the macroscopic description, one can be faced with the following seeming contradiction. The significance of the gradient terms should have increased with decreasing spatial period λ_{m}. In total, however, the components of the overall dielectric tensor $\hat{\varepsilon}_{ij}$ are known [2] to decrease exponentially with decreasing λ_{m}. Therefore, a macroscopic averaging performed for the case of crystalline media [2] would result in removal of all the short-wavelength components of the field and the material tensors, except for those corresponding to periods notably larger than the crystal lattice constants. Then a (purely practical and in no case fundamental) question arises: to what extent can the new gradient terms in (14) affect the bulk macroscopic optical properties of the medium? This is reduced to the possibility of application of a simpler CR,

$$D_i(\mathbf{r}) \approx \varepsilon_{ij}(\mathbf{r})E_j(\mathbf{r}) + \gamma^{(1)}_{ijl}(\mathbf{r})\nabla_l E_j(\mathbf{r}) + \gamma^{(2)}_{ijlm}(\mathbf{r})\nabla_l\nabla_m E_j(\mathbf{r}) + \cdots, \tag{17}$$

in which the symmetry of the material tensors is the same as in the case of homogeneous media, as a practical approximation in the phenomenological electrodynamics of macroscopically inhomogeneous media. The analysis involving additionally the wave equation shows that equation (17) can be used, instead of (14), when

$$\lambda\nabla_l\gamma^{(1)}_{ijl}(\mathbf{r}) \ll \gamma^{(1)}_{ijl}(\mathbf{r}), \tag{18}$$

and similarly for the $\gamma^{(2)}_{ijlm}(\mathbf{r})$ tensor. Formula (18) is equivalent to the condition of slow spatial variations of the material parameters ($\lambda_{\mathrm{m}} \gg \lambda$), which is justified for a large variety of solids and often for plasmas. Since it correlates well with the well-known slowly varying amplitude envelope approximation and the basic assumptions of the commonly used Jones operator calculus (see [21]), the reason for the relative success of the standard theory that neglects the material SD is obvious.

Ignoring the effect acquires even more practical significance if one remembers that formula (18) is close to the condition for applicability of the geometric optics approximation [3, 4, 21], which is best expressed in terms of the refractive indices n:

$$\lambda\nabla_l n(\mathbf{r}) \ll \overline{n(\mathbf{r})} \tag{19}$$

where $\overline{n(\mathbf{r})}$ denotes the value averaged on the light wavelength scale. When the relation (19) does not hold, we deal with an essentially inhomogeneous optical medium that strongly scatters light. Although (18) and (19) are indeed similar, unnoticed exceptions are possible for the anisotropic media, whose refractive indices are determined by a total combination of the tensor components $\varepsilon(\mathbf{r})$ and $\gamma^{(i)}(\mathbf{r})$. That is, formula (19) would remain fulfilled, despite the violation of (18), if $\varepsilon(\mathbf{r})$ and $\gamma^{(i)}(\mathbf{r})$, which are rapidly varying in space, do not contribute dominantly to the refractive indices. In other words, there might be situations when a violation of the slowly varying amplitude approximation for the anisotropic medium does not simultaneously imply large gradients $\nabla n(\mathbf{r})$, strong 'optical inhomogeneity' and the impossibility of employing the geometric optics approximation. The material SD in such non-scattering media would mainly modify a polarization state of the light waves but not their phase velocities. As shown in [23], the latter situation just occurs in crystals with the IC superstructure (see also section 4.2).

Without pretending to have achieved completeness, let us finally point out the situations in which the material SD in the non-scattering media may be of some practical importance. The appropriate conditions are easy to achieve for inhomogeneous plasmas [3]. 'Fast', though usually small in magnitude, spatial variations of the properties of condensed matter or plasmas may arise under the influence of non-uniform external fields (e.g., applied using electrodes created with the aid of modern nanotechnologies). Another example might be 'parametric' nonlinear optical effects, which consist in the 'modulation' of material parameters of a medium by

an intense (in particular, standing) electromagnetic wave, especially if that wave has a smaller length than the 'probing' wave. In this respect, liquid crystals which often manifest a very strong SD seem also to be interesting objects. It is a generally adopted approach to them to take account of a second-order SD, and so notable corrections for the material SD could be expected under some conditions. Finally, the above corrections should have a principal character in the crystalline media with long-wavelength modulated superstructure (see below).

4. Spatial dispersion in crystals with incommensurate superstructure

4.1. The microscopic constitutive relation in the Fourier domain

Now we concentrate upon the SD phenomena in crystalline materials, for which the model of a continuous medium adopted above is fitted after appropriate macroscopic averaging and the analysis of the influence of structural and superstructural periodicities [2]. Our consideration will mainly address a rather general case of dielectric crystals with IC superstructure, whereas some important results for the ordinary (non-modulated) crystals may be simultaneously obtained as a limiting case corresponding to disappearance of the IC modulation wave and the appropriate reconstruction of the set of lattice vectors. We refer the reader to the review by Cummins [24] for some introductory information on the physics of the IC phase transitions and the basic definitions. In order to make use of standard approaches adopted within the solid state theory, one should restore the translational invariance of the IC crystal, which is lost along the modulation direction during the phase transition. This may be achieved by passing formally to a superspace $(\mathbf{r}, \varphi)$, where φ denotes the phase of the IC modulation wave (see [25]). For the initial integral CR valid for a crystal inhomogeneous on the microscopic scale, the approach would mean making the parameters in (1) functions of φ (see also [26]). With formula (11), we have

$$D_i(\mathbf{r}, \varphi) = \int \epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2, \varphi) E_j(\mathbf{r}', \varphi)\, \mathrm{d}\mathbf{r}'. \tag{20}$$

Our aim is to obtain the CR that describes observable, macroscopic optical properties of the IC crystals.

On the basis of the invariance of the kernel $\epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2, \varphi)$ under the transformations imposed by the translation subgroup of the superspace symmetry group, one can write (see also appendix A)

$$\epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2, \varphi) = \epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2 + \mathbf{a}, \varphi + \mathbf{q}_{\mathrm{IC}}\mathbf{a}) \tag{21}$$

where $\mathbf{a}$ represents the translation vectors for the underlying lattice of the high-temperature parent phase and $\mathbf{q}_{\mathrm{IC}}$ the wavevector of the IC modulation. Notice that the modulation in the largest A_2BX_4 family of the IC crystals is one dimensional and occurs along the (001) direction, while $\mathbf{q}_{\mathrm{IC}} = \gamma \mathbf{a}_3^*$ and $\gamma = r/s + \delta$, where $\mathbf{a}_i^*$ are the basic reciprocal lattice vectors of the parent phase, small integer numbers r and s characterize the crystal structure of a commensurate (lock-in) phase and $\delta \ll 1$ is the incommensurability parameter [24]. In accord with (21), the translational invariance does not relate to the $\mathbf{r}_1$ variable. The additional dependence of the kernel upon $\mathbf{r}_2$ represents a periodic function that may be reduced to a Fourier series in the generalized reciprocal lattice vectors $\mathbf{h}$ of the IC phase taking the IC modulation into account ($\mathbf{h} = n_i \mathbf{a}_i^* + m\mathbf{q}_{\mathrm{IC}}$, with n_i, m being integers—see [12]). More generally, a set of reciprocal lattice vectors should be modified so as to involve any other superstructure or the periodicities due to non-uniform external fields. As a result, we have to use the following representation (cf [2], chapter 2):

$$\epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2, \varphi) = \sum_{\mathbf{h}} f_{ij}^h(\mathbf{r}_1) \mathrm{e}^{-\mathrm{i}\mathbf{h}\mathbf{r}_2} \mathrm{e}^{\mathrm{i}m\varphi}. \tag{22}$$

With (22), Fourier transforming formula (20) leads to the CR in the wavevector domain (see appendix B):

$$D_i(\mathbf{k},\varphi) = \sum_{\mathbf{h}} \hat{\varepsilon}^h_{ij}(\mathbf{k}+\tfrac{1}{2}\mathbf{h},\varphi) E_j(\mathbf{k}+\mathbf{h},\varphi) \tag{23}$$

where the Fourier components $\hat{\varepsilon}^h_{ij}$ of the microscopic dielectric permittivity tensor are defined as

$$\hat{\varepsilon}^h_{ij}(\mathbf{k}+\tfrac{1}{2}\mathbf{h},\varphi) = \mathrm{e}^{\mathrm{i}m\varphi} \int f^h_{ij}(\mathbf{r}_1)\mathrm{e}^{-\mathrm{i}(\mathbf{k}+\frac{1}{2}\mathbf{h})\mathbf{r}_1}\,\mathrm{d}\mathbf{r_1}. \tag{24}$$

The superscript h in $\hat{\varepsilon}^h_{ij}(\mathbf{k}+\frac{1}{2}\mathbf{h},\varphi)$ determines the spatial periodicity of the given Fourier component (the corresponding period is $\lambda_h = 2\pi/h$), whereas the dependence on the $\mathbf{k}+\frac{1}{2}\mathbf{h}$ argument indicates the availability of SD. The appearance of this argument originates from the definition (B.4), where the vectors $\mathbf{k}$ and $\mathbf{k}'$ enter in the same way. Therefore, the argument of the microscopic components of the dielectric tensor proves to be a 'symmetric' combination of the arguments $\mathbf{k}+\mathbf{h}$ and $\mathbf{k}$ concerned, respectively, with the electric field and the displacement in (23). This, in turn, comes from the assumption represented by formulae (11) and (12).

Since, as a rule, the SD is a weak effect and the components $f^h_{ij}(\mathbf{r}_1)$ tend rapidly to zero with increasing $\mathbf{r}_1$, we may restrict ourselves in (24) to integrating over a volume of the order of a_s^3, irrespective of $\mathbf{h}$. At the same time, the relation between the values of $f^h_{ij}(\mathbf{r}_1)$, $\hat{\varepsilon}^h_{ij}(\mathbf{k}+\frac{1}{2}\mathbf{h},\varphi)$ and $\mathbf{h}$ represents a separate problem.

The CR for the non-modulated crystals may be found from (23) under a strict condition of the absence of IC superstructure ($m = 0$):

$$D_i(\mathbf{k}) = \sum_{\mathbf{h}_0} \hat{\varepsilon}^{h_0}_{ij}(\mathbf{k}+\tfrac{1}{2}\mathbf{h}_0) E_j(\mathbf{k}+\mathbf{h}_0) \tag{25}$$

where $\mathbf{h}_0 = n_i\mathbf{a}^*_i$. Specifically the same calculations for the case of spatially non-dispersive crystal ($\epsilon_{ij}(\mathbf{r}_1,\mathbf{r}_2) = \delta(\mathbf{r}_1)\sum_{\mathbf{h}_0} f^{h_0}_{ij}\mathrm{e}^{-\mathrm{i}\mathbf{h}_0\mathbf{r}_2}$) yield

$$D_i(\mathbf{k}) = \sum_{\mathbf{h}_0} \hat{\varepsilon}^{h_0}_{ij} E_j(\mathbf{k}+\mathbf{h}_0); \tag{26}$$

i.e., the dielectric permittivity no longer depends on $\mathbf{k}+\frac{1}{2}\mathbf{h}_0$.

From (23) and (25), it is clear that the SD in a 'discretely inhomogeneous' crystalline medium can be expressed in terms of a dependence of the dielectric permittivity upon the specific combination of the light wavevector (the field SD) and the reciprocal lattice vectors (the material SD). In the case of a weak SD, the expansion of the $\hat{\varepsilon}^h_{ij}(\mathbf{k}+\frac{1}{2}\mathbf{h},\varphi)$ functions in a series of wavevector powers would immediately result in the relation between the coefficients of the field and the material SD mentioned above (see formulae (14) and (A.7), as well as the results [9, 10]). Furthermore, the wavevector dependence of the conductivity tensor derived in section 13 in [2] within a simple microscopic model for non-modulated crystals may indeed be expressed with a single argument $2\mathbf{k}+\mathbf{h}_0$, in accordance with our results[3]. The analysis of the results derived with the microscopic approach [27] confirms that the same should be true of the IC crystals.

Unlike in the present work, the microscopic dielectric Fourier components in chapter 2 of [2] appear (in our notation) as $\hat{\varepsilon}^{h_0}_{ij}(\mathbf{k})$. This is caused by the expansion of a periodic response function in the $\mathbf{r}'$ variable but not in $\frac{1}{2}(\mathbf{r}+\mathbf{r}')$ (cf formula (22)). This result contradicts the Onsager principle and disregards a possible material SD. On the other hand, the inconsistency does not matter as long as we are dealing with macroscopically homogeneous crystals, because

[3] The (quite equivalent) argument $2\mathbf{k}+\mathbf{h}$ in (24) is immediately obtained with $\alpha = 1/2$ (see also appendix A).

all the microscopic dielectric components with $\mathbf{h}_0 \neq \mathbf{0}$ can be disregarded anyway and only the 'homogeneous' components $\hat{\varepsilon}^0_{ij}(\mathbf{k})$ remain. As a matter of fact, similar inaccuracies arise in a large number of studies on the subject (e.g., $\hat{\varepsilon}_{ij}(\mathbf{k}, \mathbf{h}_0)$ in [28]; $\hat{\varepsilon}^{h_0}_{ij}(\mathbf{k}+\mathbf{h}_0)$ in [12]; see also [3, 13]). The representation of the $\hat{\varepsilon}^h_{ij}(\mathbf{k}, \mathbf{k}+\mathbf{h}, \varphi)$ kind used in [15, 26, 29] seems to be somewhat better. However, strictly speaking, such a double argument of the dielectric permittivity also contradicts the Onsager principle. In order to meet the requirements of the latter, the dielectric tensor has to be further expanded in a power series in just the 'symmetrized' combination $\mathbf{k}+(\mathbf{k}+\mathbf{h})$ of the two arguments. This point has been missed in [15, 26], though it is strictly taken into account by formulae (23) and (24).

Let us touch upon the symmetry properties of the microscopic Fourier components of the dielectric permittivity. On the basis of the obvious reformulation of (8) for the IC crystals ($\epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2, \varphi) = \epsilon_{ji}(-\mathbf{r}_1, \mathbf{r}_2, \varphi)$) and formulae (22), (24) and (B.4), one can write

$$\hat{\varepsilon}^h_{ij}(\mathbf{k}+\tfrac{1}{2}\mathbf{h}, \varphi) = \hat{\varepsilon}^h_{ji}(-\mathbf{k}-\tfrac{1}{2}\mathbf{h}, \varphi). \tag{27}$$

Moreover, the relation

$$\hat{\varepsilon}^h_{ij}(\mathbf{k}+\tfrac{1}{2}\mathbf{h}, \varphi) = [\hat{\varepsilon}^{-h}_{ij}(-\mathbf{k}-\tfrac{1}{2}\mathbf{h}, \varphi)]^* \tag{28}$$

should be fulfilled for lossless crystals. We remark in this respect that a property analogous to (27) is well known for the 'homogeneous' macroscopic dielectric component of the non-modulated crystals ($\hat{\varepsilon}^0_{ij}(\mathbf{k}) = \hat{\varepsilon}^0_{ji}(-\mathbf{k})$—see [2], chapter 1). Due to a link between the Onsager symmetry and the action of the time inversion operator $\hat{\mathbf{T}}$, the authors of [2] have treated this property as a result of the behaviour $\hat{\mathbf{T}}\mathbf{k} = -\mathbf{k}$, which becomes more evident after the identification $\hbar\mathbf{k} = \mathbf{p} = -\mathrm{i}\hbar\nabla$. However, the latter assumption has been objected to [30] as being invalid in general. The authors of [30] have stressed that the symmetry of the $\mathbf{k}$-dependent part of the dielectric tensor would be better understood as a direct consequence of relation (7).[4] The analysis of the property (27) now provides some extra reasons for accepting this point of view, since the symmetry (27) is by no means associated with the evident relation $\hat{\mathbf{T}}\mathbf{h} = \mathbf{h}$.

4.2. The mesoscopic constitutive relation for the incommensurate phases

Proceeding to the macroscopic properties of ordinary, non-modulated crystals, a common 'macroscopic approximation' is adopted, wherein all the dielectric components $\hat{\varepsilon}^h_{ij}(\mathbf{k}+\frac{1}{2}\mathbf{h}, \varphi)$ characterized by $\mathbf{h} \neq 0$ are in fact neglected (see, e.g., [2, 29]). However, the latter approach becomes inadmissably rough if we wish to describe observable optical properties of the crystals possessing IC phases [13–15]. Owing to incommensurateness of the modulation, 'ultralong-wavelength tunes' are present in their structure, which greatly exceeds the typical interatomic distances (see, e.g., the discussion by Meekes and Janner [12] for the IC phases that occur in the insulators of the A_2BX_4 group). With the aid of the four-dimensional notation [12], those spatial periodicities are expressed in terms of reciprocal lattice vectors $\mathbf{h}_I = (0; 0; n_3; m)$ having the n_3/m ratio close to $-\gamma$. They might be not too much smaller than the light wavelength or even of the same magnitude. That is why it would be natural to adopt the so-called 'mesoscopic' approximation (see [13, 14, 26, 31, 32]), which accounts for the contributions to the optical response of the IC crystal originating from the periodicities $\sim 2\pi/h_I$. As a result, the crystals in the IC phases may be regarded as being spatially inhomogeneous on the mesoscopic scale.

In the case of a plane-wave modulation region in the A_2BX_4 family materials, the vectors $\pm\mathbf{q} = \pm s\delta\mathbf{a}^*_3$ with the lowest microscopic indices n_3 and m (the relevant four-dimensional components being $(0; 0; \mp r; \pm s)$) represent the most important representatives

[4] However, it was mistakenly supposed in [30] that $\mathbf{E}^{(1)}$, $\mathbf{D}^{(1)}$ and $\mathbf{E}^{(2)}$, $\mathbf{D}^{(2)}$ relate to the points $\mathbf{r}$ and $\mathbf{r}'$, respectively.

of $\mathbf{h}_l$. The estimations give $k/q \sim 2 \times 10^{-2}$ to 10^{-1}. It has been shown in [14] that the long-wavelength modulation with the wavevector $\mathbf{q}$ essentially affects a number of physical properties of the A_2BX_4 crystals, thus justifying keeping the contributions from $\mathbf{q}$ while passing to the macroscopic parameters. Such an approach also agrees with the microscopic models which predict that the Fourier components $\hat{\varepsilon}^h_{ij}$ decrease rapidly with increasing n_3 or m (see [2], chapter 4, and the proof for the IC systems [27, 33]). Hence, we shall not discuss the known [13, 15] procedure of macroscopic averaging of equation (23) but write down straightaway the mesoscopic CR for the case of a single long-wavelength periodicity ($\lambda_m = 2\pi/q$) dominating in the structure of the IC phase:

$$D_i(\mathbf{k}, \varphi) = \varepsilon_{ij} E_j(\mathbf{k}) + \sum_{\pm\mathbf{q}} \hat{\varepsilon}^q_{ij}(\mathbf{k} + \tfrac{1}{2}\mathbf{q}, \varphi) E_j(\mathbf{k} + \mathbf{q}, \varphi) \tag{29}$$

where we have taken into account that the 'homogeneous' contributions to the first-order SD are forbidden by the inversion symmetry of A_2BX_4 crystals [12] ($\hat{\varepsilon}^0_{ij}(\mathbf{k}) \approx \varepsilon_{ij}$).

Following Golovko and Levanyuk [31], some authors believe (see, e.g., [13, 34, 35]) that the non-locality radius a_s in the IC crystals may be as large as the long-wavelength superstructure period λ_m ($\lambda_m = 2\pi/q$ in the simplest case) and so the SD should be characterized by the ratio λ_m/λ. Under such circumstances, the effect would have been huge, while the very representation of the $\hat{\varepsilon}^q_{ij}(\mathbf{k} + \frac{1}{2}\mathbf{q}, \varphi)$ function by several lowest-order terms of the series in the wavevectors is too rough. In our opinion, this questionable idea is due to identification of the non-locality scale a_s with the typical inhomogeneity scale, which does approach λ_m. Its origin is easily understood, since the said scales in fact coincide in the non-modulated crystals ($a_s \simeq a$—see, e.g., the remarks at the end of section 4.1 in [2]). However, the superstructure potential in the IC crystals is very weak with respect to the interatomic potential [24], and it is difficult to justify the assertion that the response to the electromagnetic wave should be correlated through distances of the order of $\lambda_m \gg a_s$. While doubting the possibility for the huge field SD in the IC crystals, we emphasize the dominant role of the material SD, since $\lambda_m < \lambda$ (see the quantitative evaluations [12, 13] and formula (18)).

As a result, we may expand the Fourier component $\hat{\varepsilon}^q_{ij}$ in a series and confine ourselves to considering the first-order SD only:

$$\hat{\varepsilon}^q_{ij}(\mathbf{k} + \tfrac{1}{2}\mathbf{q}, \varphi) = \varepsilon^q_{ij}(\varphi) + \mathrm{i}\gamma^q_{ijl}(\varphi)(k_l + \tfrac{1}{2}q_l) + \cdots, \tag{30}$$

where the superscript '(1)' of γ_{ijl} is dropped hereafter as unnecessary. The inverse Fourier transformation of (30) gives rise to a partial case of equation (14), the mesoscopic CR for the IC phases written in the coordinate space:

$$D_i(\mathbf{r}, \varphi) = \{\varepsilon_{ij} + \varepsilon_{ij}(\varphi + \mathbf{qr}) + \tfrac{1}{2}[\nabla_l \gamma_{ijl}(\varphi + \mathbf{qr})]\} E_j(\mathbf{r}, \varphi) + \gamma_{ijl}(\varphi + \mathbf{qr}) \nabla_l E_j(\mathbf{r}, \varphi) \tag{31}$$

where the ε_{ij} are spatially independent components, and $\varepsilon_{ij}(\varphi + \mathbf{qr})$ and $\gamma_{ijl}(\varphi + \mathbf{qr})$ periodic functions of the argument $\varphi + \mathbf{qr}$. It is worth noticing that formulae (30) and (31) agree well with the results [27] obtained in the framework of a microscopic theory. We shall use this CR for the approximate description of crystal optics of centrosymmetric IC phases in the plane-wave modulation region. Let us also stress that the criterion (18), which means the condition $\lambda_m \gg \lambda$, certainly does not hold, and the neglect of the material SD (see the approximate relation (17)) cannot be justified even as a rough approximation. Indeed, the third term in the rhs of (31) ($\sim q a_s E_0$) for fields of the type $\mathbf{E} = \mathbf{E}_0 \exp \mathrm{i}[\omega t + (k \pm q)z]$ is at least not smaller than the last one ($\sim (k \pm q) a_s E_0$). At the same time, the spatially modulated components $\varepsilon_{ij}(\varphi + \mathbf{qr})$ and $\gamma_{ijl}(\varphi + \mathbf{qr})$ proportional to a small amplitude of the modulation wave are conspicuously less than ε_{ij} and so contribute little to the refractive indices. That is why the geometric optics approximation (see formula (19)) certainly holds true; it is also quite clear from the numerous experiments. Thus, the IC crystals represent a first example of media in which material SD arises due to bulk effects and can in no case be ignored.

5. Character of light waves in crystals with incommensurate phases

We now study the influence of both the field and material SD on the light eigenwaves propagated in crystals with IC modulation. Let us consider the simplest case of light propagation along the modulation axis z in the A_2BX_4 family and neglect the modulation-induced contributions to the diagonal dielectric permittivity components ($\varepsilon_{ii}(\varphi+\mathbf{qr}) \ll \varepsilon_{ii}$). According to symmetry considerations [12], the parameters $\varepsilon_{ij}(\varphi+\mathbf{qr})$ and $\gamma_{ijl}(\varphi+\mathbf{qr})$ should be, respectively, even and odd functions of the coordinates, and

$$\begin{aligned} \varepsilon_{12}(\varphi+\mathbf{qr}) &= \varepsilon_{\mathrm{a}}\cos(\varphi+qz), \\ g_{33}(\varphi+\mathbf{qr}) &= g_{\mathrm{a}}\sin(\varphi+qz), \end{aligned} \tag{32}$$

where the gyration pseudotensor g_{ml} [20] is defined by the relation $(\omega/c)\gamma_{ijl} = e_{ijm}g_{ml}$, e_{ijm} stands for the unit antisymmetric pseudotensor of rank three and ε_{a}, g_{a} the corresponding amplitudes.

5.1. Normal waves and effective crystal optical parameters

Solving the 'wave equation' $\nabla_z^2\mathbf{E} + (\omega^2/c^2)\mathbf{D} = 0$ in the approximation linear in ε_{a}, g_{a} and using the condition of weak anisotropy in the crystal (g_{a}, ε_{a}, $\sqrt{\varepsilon_{22}} - \sqrt{\varepsilon_{11}} \ll \overline{n}$, where $\overline{n} = \sqrt{\overline{\varepsilon}} = (\varepsilon_{11}\varepsilon_{22})^{1/4}$ denotes the mean refractive index), one can obtain the expressions for the normal light waves in the medium:

$$\begin{aligned} \mathbf{E}_1(z,\varphi) &= \left\{\mathbf{e}_x + \frac{\mathbf{e}_y}{2}\left[\frac{\varepsilon_{\mathrm{a}} - g_{\mathrm{a}}(\sqrt{\varepsilon_{11}} + \frac{c}{2\omega}q)}{(\sqrt{\varepsilon_{11}} + \frac{c}{\omega}q)^2 - \varepsilon_{22}}\mathrm{e}^{\mathrm{i}(\varphi+qz)} + \frac{\varepsilon_{\mathrm{a}} + g_{\mathrm{a}}(\sqrt{\varepsilon_{11}} - \frac{c}{2\omega}q)}{(\sqrt{\varepsilon_{11}} - \frac{c}{\omega}q)^2 - \varepsilon_{22}}\mathrm{e}^{-\mathrm{i}(\varphi+qz)}\right]\right\}\mathrm{e}^{\mathrm{i}k_1 z}, \\ \mathbf{E}_2(z,\varphi) &= \left\{\frac{\mathbf{e}_x}{2}\left[\frac{\varepsilon_{\mathrm{a}} + g_{\mathrm{a}}(\sqrt{\varepsilon_{22}} + \frac{c}{2\omega}q)}{(\sqrt{\varepsilon_{22}} + \frac{c}{\omega}q)^2 - \varepsilon_{11}}\mathrm{e}^{\mathrm{i}(\varphi+qz)} + \frac{\varepsilon_{\mathrm{a}} - g_{\mathrm{a}}(\sqrt{\varepsilon_{22}} - \frac{c}{2\omega}q)}{(\sqrt{\varepsilon_{22}} - \frac{c}{\omega}q)^2 - \varepsilon_{11}}\mathrm{e}^{-\mathrm{i}(\varphi+qz)}\right] + \mathbf{e}_y\right\}\mathrm{e}^{\mathrm{i}k_1 z}. \end{aligned} \tag{33}$$

Since the perturbations of the wavevectors k_i and the resulting circular birefringence turn out to be quadratic in the modulation amplitudes, we may restrict ourselves to the zeroth approximation in what follows ($k_i = (\omega/c)\sqrt{\varepsilon_{ii}}$). Note that (33) agree with the results published previously [23, 26] and take into consideration the extra terms related to the material SD ($\sim(c/2\omega)g_{\mathrm{a}}q$).

It is useful to compare the predictions of (33) for a number of important limiting cases, conventionally regarding the q value to be arbitrary. In macroscopically homogeneous ($\lambda_{\mathrm{m}} \to \infty$) or 'slowly' ($\lambda_{\mathrm{m}} \gg \lambda_0$, with λ_0 being the light wavelength in vacuum) modulated crystals, the off-diagonal component ε_{12} of the dielectric tensor gives rise to rotation of the principal axes of the optical ellipsoid with respect to the orthorhombic x and y axes. In the case of the 'rapid' modulation ($\lambda_{\mathrm{m}} < \lambda_0$) inherent to the IC phases, ε_{12} makes the normal waves non-orthogonal ($\mathbf{E}_1\mathbf{E}_2^* \propto \varepsilon_{\mathrm{a}}(\lambda_{\mathrm{m}}/\lambda_0)^2$) and their ellipticity non-zero. The latter in fact means an apparent gyration effect. The field and the material SD lead to gyrotropy, too, while their contributions to the non-orthogonality of the normal waves cancel out under the condition $\lambda_{\mathrm{m}} \ll \lambda_0/\Delta n$ (with $\Delta n = \sqrt{\varepsilon_{22}} - \sqrt{\varepsilon_{11}}$ being the ordinary, or linear, birefringence), which is certainly satisfied for the IC phases.

This represents a rather different situation, when compared with the non-modulated crystals (cf the conclusions in [2, 23]). Thus, some crystal optical effects appear in mesoscopically inhomogeneous media, which do not have analogues in macroscopically homogeneous crystals and so are outside the corresponding classification schemes [22]. Non-uniformity of the normal waves and a spatial dependence of their polarization state represent another difficulty in solving the 'inverse problem' of crystal optics—identification of the effects

on the basis of the known normal wave polarization. According to the approach of [14, 22], the difficulty might be avoided by passing from (33) to the analysis of 'effective normal waves', whose polarization does not formally depend upon the spatial coordinates but is instead determined by the phase values $\varphi + qz$ at the boundary surfaces of a given crystal plate. Under certain circumstances, the approach may provide sample-independent results or, at least, results that are not too sensitive to the sample thickness, which are believed to be the most reasonable (see section 5.2). For the light propagation directions far from the isotropic optical axes ($g_a, \varepsilon_a \ll \Delta n$, in accordance with the experimental geometry used within the polarimetric technique HAUP—see [36]), one can obtain the following expressions for the ellipticities (χ) and the azimuths (θ) of the two effective normal waves polarized in the vicinities of the x and y axes (see also [14, 22]):

$$\begin{aligned}\chi_x &= \alpha_x^+(\cos\varphi_1 - \cos\varphi_0)\cot(\Delta/2) + \alpha_x^-(\sin\varphi_1 + \sin\varphi_0),\\ \theta_x &= -\alpha_x^-(\sin\varphi_1 - \sin\varphi_0)\cot(\Delta/2) + \alpha_x^+(\cos\varphi_1 + \cos\varphi_0),\end{aligned} \tag{34}$$

$$\begin{aligned}\chi_y &= \alpha_y^+(\cos\varphi_1 - \cos\varphi_0)\cot(\Delta/2) + \alpha_y^-(\sin\varphi_1 + \sin\varphi_0),\\ \theta_y &= \alpha_y^-(\sin\varphi_1 - \sin\varphi_0)\cot(\Delta/2) - \alpha_y^+(\cos\varphi_1 + \cos\varphi_0),\end{aligned} \tag{35}$$

where $\varphi_0 = \varphi$, $\varphi_1 = \varphi + qd$ (with d being the crystal thickness), $\Delta = (k_2 - k_1)d = (\omega/c)d\Delta n$ denotes the phase retardation due to the linear birefringence and $\alpha_x^\pm$, $\alpha_y^\pm$ are determined by the amplitudes $A_{1y}^\pm$ and $A_{2x}^\pm$, which appear as the coefficients of the terms $\exp[\pm i(\varphi + qz)]$ in formulae (33):

$$\begin{aligned}\alpha_x^+ &= -\tfrac{1}{2}(A_{2x}^+ + A_{2x}^-) \qquad & \alpha_x^- &= \tfrac{1}{2}(A_{2x}^+ - A_{2x}^-),\\ \alpha_y^+ &= -\tfrac{1}{2}(A_{1y}^+ + A_{1y}^-) \qquad & \alpha_y^- &= -\tfrac{1}{2}(A_{1y}^+ - A_{1y}^-).\end{aligned} \tag{36}$$

The parameters χ and θ would be naturally attributed, respectively, to the apparent gyration and the optical indicatrix rotation effects.

Let us again make the value q in (33) and (36) arbitrary. Taking into account, step by step, the smallness of the ratio λ_0/λ_m and the optical anisotropy parameters (the values of $g_a\Delta n$, $\varepsilon_a\Delta n$ etc) for the case of 'slow' spatial variations of the material properties, we get

$$\begin{aligned}\alpha_x^+ &= -\alpha_y^+ = \frac{g_a(\lambda_0/\lambda_m) - \varepsilon_a\Delta n}{2\overline{n}[\Delta n^2 - (\lambda_0/\lambda_m)^2]},\\ \alpha_x^- &= -\alpha_y^- = \frac{g_a\Delta n - \varepsilon_a(\lambda_0/\lambda_m)}{2\overline{n}[\Delta n^2 - (\lambda_0/\lambda_m)^2]}.\end{aligned} \tag{37}$$

This agrees with the expressions derived in [14] with the Jones calculus technique (in fact, within the slowly varying amplitude approximation). From (34), (35) and (37), one can obtain the result $\chi_y = -\chi_x$ which is well known in the gyration theory for non-modulated crystals, together with the equality $\theta_y = \theta_x$, which testifies that the θ parameter may indeed be treated as the uniquely defined rotation angle for the principal axes of the optical indicatrix ellipsoid.

When we turn to the modulation in the IC phases, it looks also to be appropriate to neglect the terms $\sim g_a\Delta n$, $\varepsilon_a\Delta n$,[5] the more so since the initial wave equation is valid under the condition $\Delta n \ll \overline{n}$. Then formulae (33) and (36) yield

$$\begin{aligned}\alpha_x^+ &\approx g_a[\overline{n}^2(\lambda_m/\lambda_0)^3 - (\lambda_m/4\lambda_0)] - (\varepsilon_a/2)(\lambda_m/\lambda_0)^2,\\ \alpha_x^- &= -\alpha_y^- \approx -\varepsilon_a\overline{n}(\lambda_m/\lambda_0)^3,\\ \alpha_y^+ &\approx g_a[-\overline{n}^2(\lambda_m/\lambda_0)^3 + (\lambda_m/4\lambda_0)] - (\varepsilon_a/2)(\lambda_m/\lambda_0)^2,\end{aligned} \tag{38}$$

where the contributions to α_x^- and α_y^- from the SD prove to be cancelled out. In compliance with the general reasoning of section 3.3 and 4.2, formulae (34), (35) and (38) predict that

[5] For transparent, non-modulated crystals, this would be equivalent to the known approximation of orthogonal normal waves (see, e.g., [2, 23]).

the values of the modulation-imposed optical parameters related to the gradient terms in (31) decrease with decreasing λ_m. A still more important consequence of these formulae is that the terms linear in the small ratio λ_m/λ_0, which originate from the material SD, can produce eigenwave ellipticity (and so gyration) at least 1000 times greater than that associated with the modulated ε_{12} component ($\sim(\lambda_m/\lambda_0)^2$) or the field SD ($\sim(\lambda_m/\lambda_0)^3$). Notice that only two the latter mechanisms for the apparent gyration have in fact been considered in the earlier studies on the subject. Furthermore, we now have $\chi_y \neq -\chi_x$ and $\theta_y \neq \theta_x$, just owing to the contributions $\sim\varepsilon_a(\lambda_m/\lambda_0)^2$ mentioned above. Therefore, the electromagnetic waves in the IC medium are not orthogonal because of both the different absolute values of their effective ellipticities and a literal non-orthogonality of the major axes of their polarization ellipses. This points once more to specific features of the crystal optics of inhomogeneous insulators (cf also the situation that arose in the absorbing modulated crystals [22]). By the way, the term 'optical indicatrix rotation' for the parameter θ [14, 23] seems to be merely conventional, in view of the latter facts.

5.2. Possibilities for detecting the material spatial dispersion effects experimentally

For more than a decade, there has been a lasting controversy concerning the possibility and the magnitude of the gyration effect observed experimentally in the centrosymmetric IC phases of the A_2BX_4 family crystals. We know that the number of relevant publications has reached at least a hundred; in particular, see the references [12–14, 26, 29, 31, 32, 34, 35, 37–42]. In view of the importance of the problem, we shall concentrate further on quantitative analysis of (34) and (38). Let us, above all, pay attention to the boundary conditions for the modulation phase $\varphi + qz$, for we would in practice have a zero value of the observed quantities χ and θ whenever the phase values φ_0 and φ_1 at the sample boundaries were not definitely fixed, irrespective of the value of the amplitude factors $\alpha^{\pm}$ [14, 41]. These questions has been discussed in the most detail in [14] (see also [13, 32, 41]). It has been assumed in [14] that $\varphi_1 = \varphi_0 + 2\pi N$ (N being an integer); i.e., an integer number of long-wavelength modulation periods λ_m should fit into the crystal length along the modulation axis. In the framework of such an assumption, one can easily arrive at a successful explanation for the hardly doubtful experimental fact of the absence of gyration in the IC phases when the light propagates along the optical axes [14]. The assumption that $\varphi_1 = \varphi_0 + (2N + 1)\pi$ looks worse in this case, although one can instead prescribe a maximum size for the gyration observed in the HAUP experiments ($\chi_x \sim g_a(\lambda_m/\lambda_0)$ whenever $\varphi_0 = 0$ or π—see formulae (34) and (38)) and predict $\theta_x = 0$. The fact is that the latter correlates on the whole with the statistics of contradictory experimental data. Irrespective of all the stated motivation concerned with the will to achieve concordance of theoretical conclusions with the experimental data, the assumptions $\varphi_0 = 0$ (or π) and $\varphi_1 = \pi$ (or 0) fundamentally agree best with the symmetry properties of the modulated functions ($\varepsilon_{ij}(\mathbf{r}) = \varepsilon_{ij}(-\mathbf{r})$ and $\gamma_{ijl}(\mathbf{r}) = -\gamma_{ijl}(-\mathbf{r})$ [12]—see figure 1).

Hence, the gyration along the essentially anisotropic directions in the IC crystal would be mainly determined by the material SD, provided that the phase factors in (34) are fixed at the boundaries and equal to the latter values. Then we have

$$\chi_x \simeq g_a(\lambda_m/2\lambda_0)\cot(\Delta/2). \tag{39}$$

It seems worth discussing the possibility of critical behaviour of χ and θ in (34), (35) and (39), when the phase retardation of the crystal sample approaches $\Delta = 2\pi N_\Delta$ (with N_Δ also being an integer). In [14], where expressions of the type of (34) occur, it has been remarked that the possibility looks rather artificial, being imposed by the preceding approximation $g_a, \varepsilon_a \ll \Delta n$

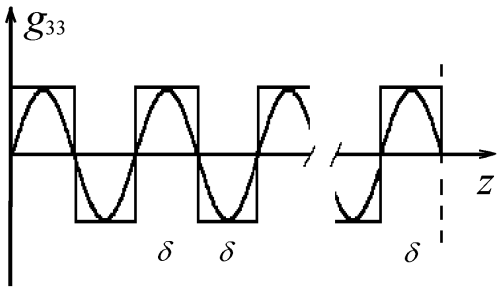

Figure 1. A schematic representation of a crystal with one-dimensionally spatially modulated gyration (see the text). The modulation phase values at the crystal boundaries are $\varphi_0 = 0$ and $\varphi_1 = \pi$, δ represents the phase retardation per modulation half-period and the light propagates along the z axis.

necessary for derivation of (34). That is, formulae (34), (35) and (39) under the given conditions would reflect nothing but the well-known [4, 5, 20, 21] tendency to a sharp increase of the normal wave ellipticity, when the propagation direction approaches the isotropic optical axes ($\Delta n \to 0$). Moreover, this by no means implies an increase in the gyration effect, since the waves become circularly polarized at $\Delta n = 0$ even for infinitely small (though non-zero) gyration parameters. Nevertheless, we find it appropriate to take much care with this problem, as a peculiarity of the type of $\cot(\Delta/2)$ in the optical parameters would evoke numerous unlikely effects dependent on the thickness ($\Delta \propto d$) of the sample under test.

First of all, the divergences $\sim a\cot(\Delta/2)$ in the temperature dependences of the polarimetric quantities directly measured with the HAUP technique (a being the small imperfection parameter of the optical equipment—see, e.g., [34, 41, 42]) have been really observed at $\Delta \to 2\pi N_\Delta$ for many crystals, and not only the modulated ones. However, no similar property has been detected in the crystal optical parameters themselves, except for the results of [43], which were later recognized by the authors themselves as erroneous [35, 39]. The exact calculation of the light eigenmode polarization parameters without any simplifying approximations represents an enormously complex task even for the non-modulated crystals (see, e.g., [2, 21]), and it is the more so for the IC crystals. Further on, we shall analyse the given problem theoretically for the similar though much simpler case of a 'slow' modulation when there exist the following favourable circumstances. First, in the case of a square waveform of the modulation (i.e., a 'discretely inhomogeneous' medium) and in the approximation related closely to the superposition principle in crystal optics [20, 21, 23], it is easy to derive the exact analytical solutions for χ with no extra assumptions about the smallness of the ratio $g_a/\Delta n$. Second, the given solutions are known to coincide almost quantitatively with those characteristic for the sine modulation wave and obtained with the differential Jones matrix calculus (see the analysis in [14]). After standard derivation [21, 32, 43] of the integral Jones matrix for a crystal with a square-wave modulation of the gyration (figure 1), one can find the ellipticity angles for the effective normal light waves:

$$\tan\chi^{\Pi}_{x,y} = \phi\cos(N+\tfrac{1}{2})B\left[\sin(N+\tfrac{1}{2})B \pm \sqrt{\sin^2(N+\tfrac{1}{2})B + \phi^2\cos^2(N+\tfrac{1}{2})B}\right]^{-1} \tag{40}$$

where

$$\begin{aligned} &\phi = \sin(\delta/2)\sin 2\chi_a/\cos(B/2),\\ &\cos B = \cos^2(\delta/2) - \cos 4\chi_a \sin^2(\delta/2). \end{aligned} \tag{41}$$

In (40) and (41), $2N + 1$ means the number of modulation half-periods that fit into the crystal thickness, $\delta = (2\pi d_m/\lambda_0)\sqrt{\Delta n^2 + (g_a/\overline{n})^2}$ the phase retardation per thickness $d_m = \lambda_m/2$ of a uniform layer corresponding to the modulation half-period (δ includes in general the contributions of both the linear (Δn) and the so-called circular [20] ($g_a/\overline{n}$) birefringences), and the ellipticity angle χ_a for the normal waves in the layer is defined by $\tan\chi_a = (g_a/\overline{n})[\Delta n + \sqrt{\Delta n^2 + (g_a/\overline{n})^2}]^{-1}$.

Formulae (40) and (41) are valid for arbitrary ratios of Δn and g_a (i.e., arbitrary propagation directions) and they predict no critical behaviour of $\chi^{\Pi}_{x,y}$. Instead, it is seen that $\sin(N + 1/2)B \to 0$ on approaching the directions of the optical axes only ($\Delta n \to 0$, $\delta \to (2\pi d_m/\lambda_0)g_a/\overline{n}$, $\chi_a \to \pm\pi/4$), when the effective normal light waves become circularly polarized ($\chi^{\Pi}_{x,y} \to \pm\pi/4$). Let us now utilize a standard method adopted for the propagation directions far from the optical axes (the HAUP-type experimental geometry; $g_a/\Delta n \ll 1$) and neglect the contributions of g_a under the radical in the formulae for δ and χ_a ($\delta \simeq (2\pi d_m/\lambda_0)\Delta n$, $(2N + 1)B \simeq (2N + 1)\delta \simeq \Delta$ and $\chi_a \simeq g_a/(2\overline{n}\Delta n)$). Putting formally $\lambda_m \ll \lambda_0$, we may pass from (40) to the relation

$$\chi^{\Pi}_x \simeq (\pi g_a/\overline{n})(\lambda_m/2\lambda_0)\cot(\Delta/2). \tag{42}$$

The latter formula correlates with the corresponding result for the 'slow' sine modulation wave obtained under the same conditions [14] (see also formulae (34) and (37)):

$$\chi^{S}_x \simeq (g_a/\overline{n})(\lambda_m/\lambda_0)\cot(\Delta/2). \tag{43}$$

It is now readily understood that the limit $\Delta n \to 0$ (or, more generally, $\Delta \to 2\pi N_\Delta$) cannot be taken correctly in the simplified formulae (42) and (43), because they are not applicable under such conditions. Eventually, the same situation happens in the case of the non-modulated crystals, for which the approximate relation $\chi_a \simeq g_a/(2\overline{n}\Delta n)$ is not applicable at $\Delta n \to 0$, either. The 'rapid' sine modulation may impose different solutions for the coefficients $\alpha^{\pm}$, while the relations (34) and (35) themselves should remain unchanged. Unfortunately, we are not in a position to get strictly the exact solutions for the 'rapid' sine modulation in the IC phases and the case of arbitrary ratios $g_a/\Delta n$, owing to extreme analytical difficulties. However, we already have many reasons to assume that the above conclusions about the '$\cot(\Delta/2)$ problem' should equally apply to (39).

Thus, the rough estimations of the apparent gyration in the IC phases may be performed while putting $\cot(\Delta/2) \sim 1$ in (39). Taking the modulation amplitude g_a as large as the order of magnitude for the gyration components in acentric crystals or somewhat smaller ($g_a \sim 10^{-6}$ to 10^{-4}, and a similar statement being true of ε_a), we obtain $\chi \sim 10^{-8}$ to 5×10^{-6} rad. The value of the ellipticity χ according to the upper limit corresponds to the sensitivity of the HAUP apparatus (see [34, 36, 41, 42]) and might, in principle, be detected experimentally. The relevant value of the effective gyration pseudotensor component ($G_{33} \simeq 2\chi\overline{n}\Delta n$ [20]) equals $G_{33} \sim 3 \times 10^{-11}$ to 10^{-7} under the reasonable conditions $\lambda_m/\lambda_0 \sim 0.02$–$0.1$, $\overline{n} \sim 1, 5$ and $\Delta n \sim 10^{-3}$–10^{-2}. It appears to be 10–10^4 times smaller than the typical gyration in the non-modulated, acentric crystals. If the boundary conditions $\varphi_1 = \varphi_0 + 2\pi N$ and $\varphi_0 = \pm\pi/2$ mentioned above hold true, formulae (34) and (38) predict that the apparent gyration is mainly determined by the modulated $\varepsilon_{12}(z)$ component rather than the material SD. Under the same conditions, we would have gyration values hardly accessible with the present-day experimental techniques ($\chi \sim 4 \times 10^{-10}$ to 3×10^{-7} rad and $G_{33} \sim 10^{-12}$ to 10^{-8}).

In conclusion, consideration of mesoscopic inhomogeneity of the A_2BX_4 group of crystals and the material SD may suggest a real mechanism for the apparent gyration effect in the IC phases characterized by inversion symmetry and, therefore, the findings of the present work may have a real impact on solving this long-standing problem. Despite very serious

discrepancies in the corresponding experimental data, the most recent studies point mainly to a small size of the effect, in agreement with our results.

Notice also that the model [37] ascribes both the optical activity and the violation of the Cauchy relations (see [37, 44]) in the A_2BX_4 family to the presence of non-zero torques, i.e. the lack of mechanical equilibrium. In this respect, subsequent electromagnetic analysis of a non-stationary incommensurately modulated medium (see section 3.3) would be useful.

6. Conclusions

In this paper, we have provided both an analytical description and a physical interpretation of the SD in microscopically and macroscopically inhomogeneous insulating media. It should be remarked that our calculations have been rather formal and involved a number of points (truncation of the series in (14) and (30), multiplying the series in (A.1), interchanging the orders of differentiation and integration in (A.7) and the same for summation and integration in (B.2)) which are allowable under the conditions of specific types of convergence for the said series. Although it would be, in principle, instructive to have a thorough mathematical proof for these, the possibility of considering the lowest-order SD as a self-dependent entity is usually justified (see [2]) for more physical than purely mathematical reasons. The relevant circumstances are provided by the relative weakness of the effect.

In the case of a weak SD, we have substantiated the quasi-local constitutive relation, starting from the general non-local integral relation and the Onsager symmetry principle, and isolated the effects termed the field SD and the material SD. As opposed to homogeneous media, an inhomogeneous, spatially dispersive medium would respond non-locally even to a spatially uniform external field.

The new gradient terms can lead to the optical gyration effect associated with finiteness of the non-locality dimension a_s with respect to the typical scales of non-uniformity λ_m of the medium. We have proven that, for the case of crystalline media, the SD should be correctly described in terms of the Fourier components of an optical frequency dielectric permittivity dependent upon a single wavevector argument, the specific combination $\mathbf{k} + \frac{1}{2}\mathbf{h}$ of the light wavevector and the generalized reciprocal lattice vectors. It is worth noting that, as far as the first-order SD is concerned, the above results, derived in a purely phenomenological way, agree with the microscopic theory [27] for the incommensurately modulated insulators, which predicts dipole–magnetodipole and dipole–quadrupole contributions to the dielectric tensor, linearly related to the mesoscopic modulation wavevector $\mathbf{q}$.

We have pointed out a first example of materials, the IC crystals of the A_2BX_4 family, whose bulk physical properties (namely, the mesoscopic periodicity with typical scales $\lambda_m \sim (20-100)a$) make the material SD important. Though our analysis hardly supports the common views regarding the possibility of a very strong field SD in the IC crystals, the calculations for the crystal optical parameters have demonstrated that the effective normal electromagnetic waves, suitable for testing experimentally, turn out to be elliptically polarized. This means that the (relatively small in size) gyration effect observed earlier in the A_2BX_4 family could be attributed to the material SD. Notice that the conclusion about a weakness of the effect does not have a general character, being rather associated with the specific structure and symmetry of the above crystals.

Finally, let us emphasize that the results of the present paper are certainly not limited to insulating condensed matter systems with two IC periodicities and should also be applicable for other inhomogeneous materials, including solid and liquid crystals under the influence of non-uniform external fields or intense electromagnetic waves, and plasmas.

Acknowledgments

The author wishes to thank Professor J F Scott for encouragement and useful suggestions. Stimulating discussions with Dr A M Shvaika and Professor I V Stasyuk are also greatly acknowledged.

Appendix A

According to (1), (11) and (12), the electric displacement function may be represented as

$$D_i(\mathbf{r}) = \int [\epsilon_{ij}(\mathbf{r}_1,\mathbf{r}) + \tfrac{1}{2}[\nabla_l \epsilon_{ij}(\mathbf{r}_1,\mathbf{r})](\mathbf{r}'-\mathbf{r})_l + \frac{1}{2!2^2}[\nabla_l\nabla_m \epsilon_{ij}(\mathbf{r}_1,\mathbf{r})](\mathbf{r}'-\mathbf{r})_l(\mathbf{r}'-\mathbf{r})_m + \cdots][E_j(\mathbf{r}) + [\nabla_l E_j(\mathbf{r})](\mathbf{r}'-\mathbf{r})_l + \frac{1}{2!}[\nabla_l\nabla_m E_j(\mathbf{r})](\mathbf{r}'-\mathbf{r})_l(\mathbf{r}'-r)_m + \cdots]\,\mathrm{d}\mathbf{r}' \tag{A.1}$$

where the action of the ∇ operator is limited by the nearest brackets. Here the expansion of the field $E_j(\mathbf{r}')$ refers to the point $\mathbf{r}' = \mathbf{r}$, while the kernel $\epsilon_{ij}(\mathbf{r}_1,\mathbf{r}_2)$ is expanded in the $\mathbf{r}_2$ argument at the point $\mathbf{b} = \mathbf{r}$, so $\mathbf{r}_2 - \mathbf{b} = (1/2)(\mathbf{r}' - \mathbf{r})$, while the derivatives mean $\nabla_{\mathbf{r}_l}\epsilon_{ij}(\mathbf{r}_1,\mathbf{r}) \equiv \nabla_{\mathbf{r}_{2l}}\epsilon_{ij}(\mathbf{r}_1,\mathbf{r}_2)|_{\mathbf{r}_2=\mathbf{r}}$ and so do not refer to the $\mathbf{r}$ variable in the argument $\mathbf{r}_1$.[6]

Dropping the coordinate indices and the arguments of the field and the response function, which are inessential in the following calculations, let us rewrite formula (A.1) in the form

$$D = \int \left[\epsilon + \frac{1}{2}(\nabla\epsilon)(r'-r) + \cdots + \frac{1}{n!2^n}(\nabla^n\epsilon)(r'-r)^n + \cdots\right] \times \left[E + (\nabla E)(r'-r) + \cdots + \frac{1}{n!}(\nabla^n E)(r'-r)^n + \cdots\right]\mathrm{d}r'. \tag{A.2}$$

After multiplying the series in (A.2) and arranging the terms in the resultant series according to increasing power of the $r'-r$ parameter, we obtain

$$D = \int \sum_{n=0}^{\infty}\left[\frac{\epsilon(r'-r)^n\nabla^n E}{n!} + \sum_{k=1}^{n}\frac{(\nabla^k\epsilon)(r'-r)^n\nabla^{n-k}E}{k!(n-k)!2^k}\right]\mathrm{d}r'. \tag{A.3}$$

Our task is to find the coefficients in an expansion analogous to that of formula (5) for the inhomogeneous medium. Besides the spatial derivatives of the field, it would also contain derivatives of the tensors defined similarly to (6), as well as 'mixed' derivatives. That is why our further transformations of (A.3) reduce to separating all possible derivatives $\nabla^q[\epsilon(r'-r)^p]$, where $q \leqslant p$. For a standard expression of the rhs of (A.3) one has

$$(\nabla^k\epsilon)(r'-r)^n = \nabla^k[\epsilon(r'-r)^n] + n\sum_{j=1}^{k}\nabla^{k-j}[(\nabla^{j-1}\epsilon)(r'-r)^{n-1}]. \tag{A.4}$$

Each term in the sum (A.4) may be reduced to the form

$$\nabla^{k-j}[(\nabla^{j-1}\epsilon)(r'-r)^{n-1}] = \sum_{i=0}^{j-1}\frac{(j-1)!(n-1)!}{i!(j-i-1)!(n-i-1)!}\nabla^{k-i-1}[\epsilon(r'-r)^{n-i-1}]. \tag{A.5}$$

[6] This becomes evident if one recalls that the $\mathbf{r}$ derivative of the tensor ε_{ij}, defined according to (6), is zero as long as the kernel depends on $\mathbf{r}$ through $\mathbf{r}_1$ only.

With (A.4) and (A.5), let us present formula (A.3) as

$$D = \int \sum_{n=0}^{\infty} \left\{ \frac{\epsilon(r'-r)^n \nabla^n E}{n!} + \sum_{k=1}^{n} \left[\frac{\nabla^k [\epsilon(r'-r)^n] \nabla^{n-k} E}{k!(n-k)! 2^k} + \sum_{j=1}^{k} \left(\sum_{i=0}^{j-1} \frac{(j-1)! n!}{i!(j-i-1)!(n-i-1)!} \frac{\nabla^{k-i-1} [\epsilon(r'-r)^{n-i-1}] \nabla^{n-k} E}{k!(n-k)! 2^k} \right) \right] \right\} \mathrm{d}r'. \tag{A.6}$$

For the fixed summation index n (i.e., in the n th approximation), a numerical coefficient for the term describing the 'zero-order SD' ϵE can be determined by putting in (A.6) $k = n$, $j = n$ and $i = n-1$. In the same way, the coefficient of the contribution to the first-order SD $\epsilon(r'-r)\nabla E$ is determined by the conditions $k = n-1$, $j = n-1$, $i = n-2$ in (A.6) etc. Therefore, the total coefficients of the contributions of all the approximations to the p th-order SD of the form $\epsilon(r'-r)^p \nabla^p E$ ($p \leqslant n$) would equal $\sum_{n=p}^{\infty} \frac{n!}{(n-p)!(p!)^2 2^{n-p}}$. Hence, the following coefficients are obtained for the lowest p: $\sum_{n=0}^{\infty} 2^{-n} = 2$ for $p = 0$; $\sum_{n=1}^{\infty} n 2^{-n+1} = 4$ for $p = 1$; and $\sum_{n=2}^{\infty} n(n-1)2^{-n} = 4$ for $p = 2$.

As a result, the expansion (A.2) reduces to the final form

$$\begin{aligned} D = \int \Bigg(& \{2\epsilon E\} + \{4\epsilon(r'-r)\nabla E + 2\nabla[\epsilon(r'-r)]E\} \\ & + \{4\epsilon(r'-r)^2\nabla^2 E + 4\nabla[\epsilon(r'-r)^2]\nabla E + \nabla^2[\epsilon(r'-r)^2]E\} + \cdots \\ & + \left\{ \sum_{n=p}^{\infty} \frac{n![\epsilon(r'-r)^p]\nabla^p E}{(n-p)!(p!)^2 2^{n-p}} + \cdots + \sum_{n=p}^{\infty} \frac{n!\nabla^p[\epsilon(r'-r)^p]E}{(n-p)!(p!)^2 2^n} \right\} + \cdots \\ & + \left\{ \frac{1}{n!}\epsilon(r'-r)^n \nabla^n E + \cdots + \frac{1}{n!2^n}\nabla^n[\epsilon(r'-r)^n]E \right\} + \cdots \Bigg) \mathrm{d}r' \end{aligned} \tag{A.7}$$

where each curly brace combines the terms associated with the SD of a common order. The numerical coefficients of the lowest $(r'-r)$-order terms of interest to us would approach with increasing accuracy the values given by (A.7) as the approximation used became higher (i.e., with increasing n). Retaining only the lowest-order SD terms in (A.7) in the limit $n \to \infty$, writing out the coordinate indices and the arguments of all the functions again, and interchanging the orders of differentiation and integration, we arrive at formulae (14) and (15) of this work.

We finally stress that the numerical coefficients $1/2^n$, which appear in the expansion (A.1) and turn out to be crucial in the following discussion, have nothing to do with the particular definition $\beta = 1/2$ used by us. In a general case of $\mathbf{r}_2 = \beta(\mathbf{r}+\mathbf{r}')$, the kernel is expanded at the point $\mathbf{b} = 2\beta\mathbf{r}$ and we nevertheless have $\epsilon(\mathbf{r}_1, \mathbf{r}_2) = \epsilon(\mathbf{r}_1, 2\beta\mathbf{r}) + \frac{1}{2}[\nabla_{\mathbf{r}}\epsilon(\mathbf{r}_1, 2\beta\mathbf{r})]\cdot(\mathbf{r}'-\mathbf{r}) + \cdots$, where the coordinate indices are dropped for conciseness. However, the handling of the values $\beta \neq 1/2$ involves some weak points. First, the kernel and the field in the rhs of (A.1) are defined at different points ($2\beta\mathbf{r}$ and $\mathbf{r}$, respectively). Second, for periodic crystalline media, the periodicity expressed in terms of $\mathbf{r}_2$ gets altered, compared to the periodicity in terms of $\mathbf{r}$ and $\mathbf{r}'$ coordinates ($\epsilon_{ij}(\mathbf{r}, \mathbf{r}') = \epsilon_{ij}(\mathbf{r}+\mathbf{a}, \mathbf{r}'+\mathbf{a})$ and $\epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2) = \epsilon_{ij}(\mathbf{r}_1, \mathbf{r}_2 + 2\mathbf{a})$ for $\beta = 1$—see section 4.1). In this respect, the choice of the α constant in the definition $\mathbf{r}_1 = \alpha(\mathbf{r} - \mathbf{r}')$ is not so limited.

It is also worth noticing that the construction of an expansion such as (14), which violates the assumption concerning the additional dependence of the response function on $\beta(\mathbf{r}+\mathbf{r}')$, would have met with insurmountable difficulties. If, for example, the kernel $\epsilon_{ij}(\mathbf{r}-\mathbf{r}', \mathbf{r}')$ were used, then on the basis of $\epsilon_{ij}(\mathbf{r}_1, \mathbf{r}') = \epsilon_{ij}(\mathbf{r}_1, \mathbf{r}) + \nabla_l[\epsilon_{ij}(\mathbf{r}_1, \mathbf{r})](\mathbf{r}' - \mathbf{r})_l + \cdots$, we would obtain a relation of the type of (A.7), in which, however, the numerical coefficients

of $\epsilon(r'-r)^p\nabla^p E$ form the divergent series $\sum_{n=p}^{\infty} n![(n-p)!(p!)^2]^{-1}$. As a consequence, the coefficients appearing in the definitions of the material tensors $\varepsilon(\mathbf{r})$ and $\gamma^{(i)}(\mathbf{r})$ (see formulae (15)) would have changed every time on raising the accuracy of the expansion (increasing n), thus evidencing that the corresponding representation of the product of the series for the kernel and the field (see (A.1) and (A.2)) in a form similar to formula (14) is incorrect. Even if we did not care about the 'divergence problems' and confined ourselves to a low-order (second-order, third-order etc) approximation for the $\varepsilon_{ij}(\mathbf{r})$, $\gamma^{(1)}_{ijl}(\mathbf{r})$ and $\gamma^{(2)}_{ijlm}(\mathbf{r})$ tensors, the corresponding 'quasi-local' CR would still not have met the requirements of the Onsager principle (see section 3.2).

Appendix B

Standard Fourier transformation of (20) gives

$$D_i(\mathbf{k},\varphi) = \int \epsilon_{ij}(\mathbf{k},\mathbf{k}',\varphi)E_j(\mathbf{k}',\varphi)\,\mathrm{d}\mathbf{k}' \tag{B.1}$$

where

$$\epsilon_{ij}(\mathbf{k},\mathbf{k}',\varphi) = \frac{1}{(2\pi)^3}\int\int \epsilon_{ij}(\mathbf{r}_1,\mathbf{r}_2,\varphi)\mathrm{e}^{\mathrm{i}(\mathbf{k}'\mathbf{r}'-\mathbf{k}\mathbf{r})}\,\mathrm{d}\mathbf{r}\,\mathrm{d}\mathbf{r}'. \tag{B.2}$$

Substituting (22) into (B.2) and changing the integration variables, one gets

$$\epsilon_{ij}(\mathbf{k},\mathbf{k}',\varphi) = \sum_{\mathbf{h}} \hat{\varepsilon}^h_{ij}(\tfrac{1}{2}(\mathbf{k}+\mathbf{k}'))\delta(\mathbf{k}'-\mathbf{k}-\mathbf{h})\mathrm{e}^{\mathrm{i}m\varphi} \tag{B.3}$$

where

$$\hat{\varepsilon}^h_{ij}(\tfrac{1}{2}(\mathbf{k}+\mathbf{k}')) = \int f^h_{ij}(\mathbf{r}_1)\mathrm{e}^{-\frac{\mathrm{i}}{2}(\mathbf{k}+\mathbf{k}')\mathbf{r}_1}\,\mathrm{d}\mathbf{r}_1. \tag{B.4}$$

Then formula (23) of section 4.1 may be readily obtained from (B.1) and (B.3).

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